

A Satellite-based Assessment of Trans-Pacific Transport of Pollution Aerosol

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Popular Summary

Air pollution has no boundary. Three decades of satellite images provide mounting evidence of the intercontinental and hemispheric transport of aerosols, such as the unraveling of “Arctic haze”, the sweep of Saharan dust over the Atlantic Ocean, and the detection of Asian pollution and dust in North America. Because of poor quality of early-generation satellite observations, however, these impressive images only provide qualitative or semi-quantitative estimates of long-range transport of aerosols, which is not adequate to address important implications of continental scale transport of aerosol for air quality, climate change, atmospheric chemistry, and ocean biology applications.

What can satellite data tell us about the long range transport of pollution beyond the pretty pictures? As a result of the much improved measurement accuracy and enhanced new capabilities of satellite sensors such as those onboard the A-Train, quantitative assessments of aerosol long-range transport from measurements just became feasible very recently. Such measurement-based assessments can augment intensive field campaigns by expanding temporal and spatial scales and also serve as constraints for model simulations.

In this study we estimated interannual and seasonal variations of mass fluxes of pollution aerosol exported from East Asia to the North Pacific basin (East Asia outflow) and imported to North America (North America inflow) based on satellite data supplemented by *in situ* measurements. The method takes advantage of MODIS high-accuracy measurements of aerosol optical depths over ocean, fine- and coarse-mode separately, in combination with satellite measurements of vertical profiles of aerosol and water vapor respectively from GLAS and AIRS, and *in situ* measurements of comprehensive aerosol chemistry and physics in the regions.

Our calculations show that about 18 Tg pollution aerosol is exported from East Asia to the northwestern Pacific Ocean each year, of which about 25% reaches the west coast of North America. The pollution fluxes are largest in spring and smallest in summer, which is mainly controlled by Asian monsoon circulations. During 2002-2005 the strongest export and import of particulate pollution occurred in 2003, due largely to record intense and persistent biomass fires from Siberia in summer and spring. Model

simulations of two NASA aerosol models, namely GOCART and GMI, agree quite well with the satellite-based assessment of annual pollution fluxes.

Such measurement-based estimate of trans-Pacific transport is critical to the assessment of how pollution from Asia influences climate and air quality in North America. It could also play a significant role in formulating effective control strategies in North America for combating air quality degradation and climate change. This study is the first large-scale *measurement-based* estimate of intercontinental pollution transport. Air pollution is a global problem and the satellite-based approach developed in this study can be adapted and extended to study the intercontinental transport of pollution and dust in other regions.

A Satellite-based Assessment of Trans-Pacific Transport of Pollution Aerosol

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Abstract: It has been well documented that pollution aerosol and dust from East Asia can transport across the North Pacific basin, reaching North America and beyond. Such intercontinental transport extends the impact of aerosols for climate change, air quality, atmospheric chemistry, and ocean biology from local and regional scales to hemispheric and global scales. Long term, measurement-based studies are necessary to adequately assess the implications of these wider impacts. A satellite-based assessment can augment intensive field campaigns by expanding temporal and spatial scales and also serve as constraints for model simulations. Satellite imagers have been providing a wealth of evidence for the intercontinental transport of aerosols for more than two decades. Quantitative assessments, however, became feasible only recently as a result of the much improved measurement accuracy and enhanced new capabilities of satellite sensors. In this study, we generated a 4-year (2002 to 2005) climatology of optical depth for pollution aerosol (defined as a mixture of aerosols from urban/industrial pollution and biomass burning in this study) over the North Pacific from MODerate resolution Imaging Spectro-radiometer (MODIS) observations of fine- and coarse-mode aerosol optical depths. The pollution aerosol mass loading and fluxes were then calculated using measurements of the dependence of aerosol mass extinction efficiency on relative humidity and of aerosol vertical distributions from field campaigns and available satellite observations in the region. We estimated that about 18 Tg/year pollution aerosol is exported from East Asia to the northwestern Pacific Ocean, of which about 25% reaches the west coast of North America. The pollution fluxes are largest in spring and smallest in summer. For the period we have examined the strongest export and import of pollution particulates occurred in 2003, due largely to record intense Eurasia wildfires in spring and summer. The overall uncertainty of pollution fluxes is estimated at about 80%. A reduction of uncertainty can be achieved with a better characterization of pollution aerosol through integrating emerging A-Train measurements. Simulations by the Goddard Chemistry Aerosol Radiation and Transport (GOCART) and Global Modeling Initiative (GMI) models agree quite well with the satellite-based estimates of annual and latitude-integrated fluxes, with larger model-satellite differences in latitudinal variations of fluxes.

1. Introduction

1.1. Asian pollution and intercontinental transport

East Asia is an important source region of a variety of natural and anthropogenic aerosols, also called particulate matter (PM). China, the world's most populous country has been undergoing persistent rapid industrialization and urbanization and the burgeoning expansion of automobile usage over the last two decades. The nation relies heavily on coal burning to meet ever-increasing energy needs (*Liu and Diamond, 2005*) and consumes large amounts of low efficiency biofuels (*Streets and Aunan, 2005*). China has more than doubled its emissions in recent decades to quickly become one of the largest emitters of aerosols and aerosol precursors in the world (*Ohara et al., 2007; Richter et al., 2005*) and is the largest contributor of sulfur dioxide (SO₂) to the atmosphere. The Chinese emission of carbonaceous aerosols is nearly the sum of North America and Europe emissions (*Bond et al., 2004*) and the observed concentrations for carbonaceous aerosols in mega-cities are 1~3 orders of magnitude higher than those reported from developed countries (*Wang et al., 2006*). While Chinese emissions of SO₂, carbonaceous aerosols and CO showed a decreasing trend during 1996-2000, emissions for these species have since been increasing (e.g., *Carmichael et al., 2002; Ohara et al., 2007*). The increased emission of pollutants in East Asia is projected to continue in the coming decades, although an accurate projection of the rate of increase is complicated by uncertainties associated with future economic growth, air pollution control policy, and bottom-up emission inventory itself, among others (*Carmichael et al., 2002; Akimoto et al., 2006; Ohara et al., 2007*). Dust emissions in the northwestern plains of China are comparable with that of the Saharan dust. Dust storms frequently blanket a large area of northern China in the spring season and have become

increasingly intense over the past few years after a decreasing trend from 1969-1999 (*Zhang et al.*, 2003).

Such large emissions of aerosols may have brought about substantial impacts to China's climate (*Huang et al.*, 2006), hydrological cycle (*Menon et al.*, 2002), solar radiation reaching the surface (*Qian et al.*, 2007), crop yields (*Chameides et al.*, 1999), and human health (*Xu et al.*, 1998). Furthermore, Asian aerosol could impose far-reaching environmental impacts at regional, hemispherical and even global scales, because of mounting evidence for intercontinental and even hemispheric transport provided by long-term surface monitoring networks, in-situ measurements from intensive field campaigns, and satellite observations backed by model simulations. Dust storms and industrial emissions have long been observed to influence the North Pacific (*Duce et al.*, 1980), to contribute to particulate matter level on the western boundary of North America (*Andreae et al.*, 1988; *Jaffe et al.*, 1999; *Husar et al.*, 2001; *Wilkening et al.*, 2000; *McKendry et al.*, 2001; *Jaffe et al.*, 2005) and on the East Coast after traversing the continent (*Biscaye et al.*, 2000; *VanCuren and Cahill*, 2002), and to even affect Europe (*Stohl et al.*, 2007; *Grousset et al.*, 2003). Model simulations also suggest substantial trans-Pacific transport of aerosols (*Chin et al.*, 2004, 2007; *Heald et al.*, 2006; *Hadley et al.*, 2007).

East Asia is one of the main warm conveyor belts (WCB) inflow regions, where airstreams transport the boundary layer air into the upper troposphere on time scales of 1-2 days (*Stohl*, 2001; *Eckhardt et al.*, 2004). Other vertical lifting mechanisms include frontal and postfrontal convection, orographic lifting, and the atmospheric boundary layer (ABL) turbulent

mixing. As a result, emissions from Asia can experience rapid vertical transport (*Stohl et al.*, 2002). Pollutants lifted to the free troposphere travel initially poleward and eastward by mid-latitude storm tracks, turn towards the equator thereafter, and then reach mainly the middle and upper troposphere of the northeastern Pacific. Such high-level transpacific transport takes about a week (*Holzer et al.*, 2005). The polluted Asian air over the Northeastern Pacific can then be transported downward to influence the North American ABL through subsidence associated with subtropical Pacific highs (*Heald et al.*, 2003) and through meso-scale mountain circulations in Western North America (*McKendry et al.*, 2001). Air pollutants can also be transported across the North Pacific at low altitudes through ABL outflow associated with cold fronts (*Liang et al.*, 2004; *Liu et al.*, 2003), with transit times 1-2 weeks longer than the high-level trans-Pacific transport (*Holzer et al.*, 2005). Specific mechanisms responsible for trans-Pacific transport depend strongly on season and location (*Eckhardt et al.*, 2004; *Liang et al.*, 2004; *Holzer and Hall*, 2007; *Dickerson et al.*, 2007). This in combination with variations in pollutant emissions could result in pronounced seasonal and interannual variations of pollution outflow and inflow.

The regional and global impacts of Asian aerosols are largely unknown but the implications are quite powerful. A tracer simulation suggests that Asian CO emissions with an assumed atmospheric lifetime of 5 days (comparable to lifetimes of tropospheric aerosol) could contribute to the total CO column over North America by about 30% (*Stohl et al.*, 2002). Because particulates with a diameter less than 2.5 μm (PM_{2.5}), can efficiently penetrate into the lungs and cause cardiovascular and respiratory diseases, increasingly stringent standards for PM_{2.5} have been enforced by the U.S. Environmental Protection Agency (EPA). While most pollution sources are local, the trans-boundary and intercontinental transport of PM_{2.5} from Asia

may compromise efforts of some western U.S. states to attain air quality goals through domestic and regional emission controls (e.g., *Heald et al.*, 2006; *Chin et al.*, 2007; *Fairlie et al.*, 2007). Aerosols can perturb the radiative budget of the earth-atmosphere system directly through scattering and absorbing radiation and indirectly through altering cloud formation and cloud properties. The deposition of soot in snow and ice can also reduce surface reflectance and hence perturb the surface energy budget (*Hansen and Nazarenko*, 2004). Linked through these processes, aerosols can thereby affect atmospheric circulations and precipitation. It has been suggested that an increase of particulate air pollution may reduce orographic precipitation in the mountain ranges of the western United States (*Rosenfeld and Givati*, 2006; *Jirak and Cotton*, 2006), causing a significant shortage of water resources in the western states. Asian dust particles have been observed to affect the formation of clouds over the western US (*Sassen*, 2002). A satellite-detected intensification of the North Pacific mid-latitude storm tracks has been associated with an increase of aerosol emission in Asia in the last two decades (*Zhang et al.*, 2007). This suggests a possible feedback influence of long-range transport of aerosols, because the North Pacific storm tracks effectively transport pollution and dust from East Asia to North America.

1.2. A satellite perspective

A better understanding of aerosol transport, transformation, and distribution is required to assess a variety of aerosol impacts on a wide range of scales and formulate an effective strategy for combating air pollution in the U.S. Due to the large emissions and rate of increase in Asia, the potential impacts on the environment are relatively easy to detect and can be studied over a large dynamic range. As such, in the past two decades, the aerosol related scientific and

environmental issues in the North Pacific basin have been a focus of several large scale international experiments, including the Western Pacific Exploratory Missions (PEM-West A and B) (*Hoell et al.*, 1996, 1997), Aerosol Characterization Experiment-Asia (ACE-Asia) (*Huebert et al.*, 2003), TRANsport and Chemical Evolution over the Pacific (TRACE-P) (*Jacob et al.*, 2003), Photochemical Ozone Budget of the Eastern North Pacific Atmosphere (PHOBEA) (*Bertschi, et al.*, 2004), Intercontinental Transport and Chemical Transformation (ITCT 2K2), and Pacific Exploration of Asian Continental Emission (PEACE) (*Parrish et al.*, 2004), Intercontinental Chemical Transport Experiment – Phase B (INTEX-B), among others. During some of these intensive field campaigns, aerosols have been studied in great detail using in-situ and remote sensing observations of physical and chemical properties from multiple platforms such as aircraft, ship, and ground networks and model simulations as well. These missions have been providing comprehensive snapshots of regional aerosols that can be intercompared to help understand the complex interactions of aerosols within atmospheric systems.

For all of their advantages, field campaigns are inherently limited by their relatively short duration and small spatial coverage. Therefore they are not adequate to assess the temporal and spatial variations of trans-Pacific transport of aerosols. Satellite remote sensing of aerosols can augment these missions by expanding temporal and spatial scales to generate measurement based estimates of aerosol intercontinental transport. In addition, these estimates can serve as constraints for model simulations. Satellite imagers not designed specifically for aerosol research have been providing a wealth of evidence for intercontinental and hemispheric transport of aerosols in a qualitative or semi-quantitative manner for nearly three decades (e.g., *Lyons et al.*, 1978; *Chung*, 1986; *Herman et al.*, 1997). Quantitative estimates of aerosol mass and transport were also attempted (*Fraser*, 1976; *Fraser et al.*, 1984). However these early estimates were

subject to large uncertainties because of poor accuracy of early satellite measurements. Quantitative assessments of aerosol intercontinental transport became feasible only recently as a result of the much improved measurement accuracy and enhanced new capabilities of satellite sensors (King et al., 1999; Kaufman et al., 2002), as well as a constellation of multiple satellites (such as “A-Train”) with complementary measurement capabilities. Although current and near-future satellite sensors can not directly measure aerosol composition, measurements can be used to categorize aerosol types in terms of particle size (i.e., fine vs coarse), shape (i.e., spherical vs non-spherical), and absorption (i.e., scattering vs absorbing) because of implementation of multi-wavelength, multi-angle, and polarization measurements (Higurashi and Nakajima, 2002; Tanré et al., 2001). Algorithms have been developed to quantitatively estimate the optical depth of pollution aerosol and dust (e.g., Kaufman et al., 2005a, b). The new capabilities associated with these passive sensors are being further enhanced by emerging lidar measurements in recent years that are providing essential information of aerosol vertical distributions (Spinhirne et al., 2005; Winker et al., 2003). It is currently feasible to develop a satellite-based approach supplemented by in situ measurements and model simulations to quantify aerosol intercontinental transport.

In this study we make our first attempt at this approach by estimating the mass fluxes of pollution aerosol exported from East Asia to the North Pacific basin and imported to North America based on satellite data. For this purpose, the satellite observed aerosol is categorized into three generic types based on its sources, i.e., maritime aerosol, mineral dust, and pollution aerosol. Here *pollution aerosol* is considered to be a mixture of aerosols from urban/industrial pollution and biomass burning, which is predominately in sub-micron size or fine-mode. All biomass burning is assumed to be man made here. The method takes advantage of the Moderate

resolution Imaging Spectroradiometer (MODIS) high-accuracy measurements of aerosol optical depths over ocean, fine- and coarse-mode separately, in combination with satellite measurements of aerosol vertical profiles from the Geoscience Laser Altimeter System (GLAS), and in situ measurements of comprehensive aerosol chemistry and physics from intensive field campaigns in the regions. The estimated aerosol import and export fluxes are compared with the Goddard Chemistry Aerosol Radiation Transport (GOCART) and Global Modeling Initiative (GMI) simulations. The satellite observations of aerosol and aerosol models are described in Section 2. In Section 3, an approach to deriving pollution fluxes from satellites is presented. Seasonal and interannual variations of the satellite-estimated export and import of pollution aerosol are discussed and compared with the model simulations in Section 4. Uncertainties associated with the flux estimate and potential of reducing uncertainties by integrating the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) and other A-Train observations are also discussed. Major results are summarized in Section 5.

2. Description of Aerosol Data and Models

2.1. MODIS aerosol optical depths

MODIS aboard both Terra and Aqua is making near global daily observations of atmospheric aerosols since February 2000 (Terra) and July 2002 (Aqua). MODIS uses seven wavelength channels (between 0.47 and 2.13 μm) to retrieve aerosol properties over cloud and surface-screened areas. Separate algorithms are implemented over land and ocean (*Kaufman et al., 1997; Tanré et al., 1997; Remer et al., 2005; Levy et al., 2007*). MODIS products are widely used by the community for a variety of purposes, including descriptions of the regional, seasonal and global distribution of aerosols and their relationship to other pollutants (*Yu et al., 2003; Chin*

et al., 2004), studies of the effect of aerosol on atmospheric chemistry and local air pollution (Engel-Cox *et al.*, 2004; Al-Saadi *et al.*, 2005), measurements of the aerosol radiative forcing of climate (Yu *et al.*, 2004; Remer and Kaufman, 2006), studies of the aerosol interaction with the meteorological field and with clouds (Koren *et al.*, 2004; Kaufman *et al.*, 2005d), and synergy with other sensors to enhance standard MODIS retrievals and offer new standalone products (Kaufman *et al.*, 2003a,b; León *et al.*, 2003).

Because of its wide spectral range over ocean, MODIS has the unique capability to retrieve not only spectral aerosol optical depth (τ) at seven wavelengths from 0.47 to 2.13 μm with great accuracy, i.e., $\pm 0.03 \pm 0.05\tau$ (Remer *et al.*, 2002, 2005), but also the quantitative aerosol size parameters (e.g., effective radius and fine-mode fraction) (Kaufman *et al.*, 2002; Remer *et al.*, 2005; Kleidman *et al.*, 2005). The fine-mode fraction (FMF) is a measure of the contribution of fine-mode aerosols to the optical depth. Because anthropogenic aerosols are predominately fine-mode or in submicron range, FMF can be used as a tool for separating anthropogenic aerosol from dust, as demonstrated in some previous studies. Kaufman *et al.* (2005a) developed an algorithm that uses MODIS/Terra aerosol optical depth (AOD) and FMF to distinguish dust from pollution and maritime aerosols and evaluate the column concentration, transport and deposition of dust over the Atlantic Ocean. The results confirm seasonal variations of magnitude and location of African dust transport across the Atlantic Ocean. In particular, the estimated deposition of 50 Tg dust in the Amazon, much larger than previous estimates, may explain the paradox between the need of nutrition by the Amazon forest and the source of the nutrition. A similar algorithm has been employed to estimate anthropogenic and natural components of aerosols (Kaufman *et al.*, 2005b), which has since inspired the community to

further explore the use of satellites to quantify aerosol radiative forcing by anthropogenic aerosol (e.g., *Anderson et al.*, 2005a, *Bellouin et al.*, 2005; *Christopher et al.*, 2006; *Yu et al.*, 2006).

2.2. ICESat and CALIPSO aerosol profiles

GLAS was launched in early 2003 aboard ICESat to become the first polar orbiting satellite lidar with the ranging capability to serve as both a precision surface elevation altimeter and an atmospheric lidar (*Spinhirne et al.*, 2005). CALIPSO is a joint U.S. and French satellite mission with an expected 3 year lifetime (<http://www-calipso.larc.nasa.gov/>). Launched on April 28, 2006, CALIPSO combines an active lidar instrument (CALIOP) with passive infrared and visible imagers (*Winker et al.*, 2003). GLAS and CALIOP both use 532 nm and 1064 nm wavelengths to probe the vertical structure and properties of thin clouds and aerosols with a resolution of ~60 m vertically and ~10 km along track. The 532 nm wavelength of CALIOP also has a capability of detecting polarization, which can facilitate the separation of non-spherical dust from spherical particles. These satellite sensors cover the globe between ~80°N and ~80°S latitudes, with an orbital repeating cycle of about 16 days. Although they are nadir track only sensors, useful global distributions can emerge for monthly compilations involving hundreds of orbits.

Since February 2003, GLAS has operated during specific time segments of about 4 to 8 weeks duration due to the limited laser lifetime. Although limited in time coverage, the GLAS data provide, for the first time, the vertical distributions of aerosols backscatter at 532 nm up to a 40 km height on a global scale (*Spinhirne et al.*, 2005). While similar products will be derived from CALIPSO measurements that are expected to be continuous in temporal coverage, the full

calibrations and aerosol-cloud separation have not yet been implemented at the time of conducting this study. In this study, we will examine GLAS data only and discuss the potential of CALIPSO data for better determining the major transport heights of pollution plumes and a reduction of uncertainty associated with flux estimates in Section 4.3.

2.3. GOCART and GMI aerosol simulations

The global model GOCART is driven by assimilated meteorological fields from the Goddard Earth Observing System Data Assimilation System (*GEOS DAS*) (*Chin et al.*, 2002). GOCART simulates the major aerosol types: sulfate, dust, black carbon, organic carbon, and sea-salt. Emissions from anthropogenic, biomass burning, biogenic, and volcanic sources and wind-blown dust and sea-salt are included in the model. Processes represented in GOCART are chemistry, convection, advection, boundary layer mixing, dry and wet deposition, gravitational settling, and hygroscopic growth of aerosol particles. Details of GOCART and evaluation of its results against observations are documented in a number of publications (e.g., *Chin et al.*, 2000a, 2000b, 2002, 2004; Ginoux et al., 2001, 2004).

The Global Modeling Initiative (*GMI*) was established to understand and reduce uncertainties in assessing the impact of various natural and anthropogenic perturbations on atmospheric composition and chemistry. The GMI aerosol module, adapted from the University of Michigan/LLNL IMPACT model (*Liu et al.*, 2007), is similar to the GOCART model in such aspects as fossil fuel emissions of SO₂, BC, and OC, and the advection core. On the other hand, the two models differ in volcanic emissions, aqueous phase reactions, vertical diffusion, and dry deposition and wet scavenging schemes, among others. As such GOCART and GMI simulated

mass and flux of pollution aerosol can be different both in the horizontal and vertical, though both models are driven by the same meteorological fields from GEOS-4 in this study.

For a comparison with MODIS-based estimates of pollution flux, we use a sum of model simulated black carbon, organic matters, and sulfate as a proxy for pollution aerosols. While this sum is higher than the modeled “pollution” aerosols since there are natural components of sulfate and organic matter, the bias could be partially offset by an exclusion of nitrate and ammonium in the models that is largely anthropogenic. Note also that for the MODIS estimate we have assumed all biomass burning smoke is man-made pollution, which is somewhat consistent with the pollution proxy assumption made for the models.

3. Approaches to estimating pollution fluxes from satellites

In this study we extend the method and analysis in Kaufman et al. (2005a, b) to estimate fluxes of pollution aerosol exported from East Asia and imported to North America. A three-step method integrates datasets from multiple satellite sensors and *in situ* measurements from field campaigns to generate a four-year (2002-2005) climatology of trans-Pacific pollution aerosol transport, as discussed in the following.

3.1. Deriving pollution aerosol optical depth from MODIS retrievals

We use the method developed by Kaufman et al. (2005a,b) to derive pollution aerosol optical depth (τ_p) according to:

$$\tau_p = \frac{(f - f_d)\tau - (f_m - f_d)\tau_m}{(f_p - f_d)} \quad (1)$$

where τ and f represents aerosol optical depth and fine-mode fraction respectively, both at 550 nm. Subscripts p, d, and m denote pollution, dust, and maritime aerosol, respectively. Here we assume that volcanic aerosols are negligible. Such assumptions may bias our estimate of pollution optical depth to a high value in years following major volcano eruptions. We applied the method to Collection 4, Level 3 daily MODIS/Terra aerosol data (at $1^\circ \times 1^\circ$ resolution) so that empirical coefficients in (1), f_m , f_d , f_p , and τ_m derived in Kaufman et al. (2005a, b) are valid.

We modify the method developed in Kaufman et al. (2005a, b) by taking account of seasonal and spatial variations of fine-mode fraction for maritime aerosol (f_m). Over remote oceans, boundary layer aerosols consist of sea salt particle generated from bursting bubbles, sulfate from oxidation of dimethyl sulfide (DMS), and organic matter. While DMS-oxidized sulfates and organic particles are in the sub-micron size range, the sea salt aerosols have much broader size distributions (*Bates et al.*, 2001) and the fine-mode sea salt is a significant contributor to sea salt optical depth and also an important component of fine-mode maritime aerosol optical depth. These marine-generated fine-mode aerosols should depend on a variety of atmospheric and oceanic parameters, such as ocean color, sea-surface temperature (SST), near-surface wind speed, and atmospheric oxidizing capacity. This complexity gives rise to large seasonal and geographical variations in the maritime FMF (ranging from 0.2 to 0.8), as evidenced consistently in a MODIS-derived proxy for maritime aerosol (e.g., AOD less than 0.1) and GOCART and GMI simulations of purely maritime aerosol (Yu et al., *Seasonal and geographical variations of maritime aerosol fine-mode fraction and implication for deriving MODIS anthropogenic aerosol*, in preparation). A use of the seasonally and geographically varying maritime FMF decreases the pollution aerosol optical depth by as much as 30% in

coastal areas of the North Pacific and in summer, in comparison to that using the constant f_m (Kaufman et al., 2005a,b).

3.2. Determining major transport heights of pollution aerosol

To transform aerosol optical depth to aerosol mass loading and flux, major transport heights of pollution aerosol are needed to determine the relative humidity and wind speed representative of satellite observed pollution plumes. Passive sensors like MODIS acquire columnar aerosol properties which give essentially no information on vertical distributions of aerosol. In this study we determine the transport heights of pollution aerosol based on aircraft measurements in the region, available surface and satellite lidar observations, and model simulations.

Observations over the northwestern Pacific during ACE-Asia, TRACE-P, and PEM-West B indicate multiple pollution layers from the surface up to the middle troposphere with a significant fraction of pollution aerosol mass above the ABL (e.g., Bahreini et al., 2003). For example, SO_4^{2-} and NH_4^+ profiles over Northwestern Pacific indicate that about one third of sulfate mass is above 2 km (Dibb et al., 2003; Chin et al., 2003). Carbonaceous aerosols were even more concentrated than sulfate in the upper troposphere (Huebert et al., 2004). A global 3-D chemical transport model showed that the trans-Pacific transport of Asian anthropogenic aerosols takes place mainly in 900-700 hpa layer because of scavenging during transport either in the boundary layer or during lifting to the upper troposphere (Heald et al., 2006). Measurements over the Northeastern Pacific Ocean and Western North America indicate persistent Asian continental influences in the lower free troposphere (VanCuren et al., 2005). While Siberian

boreal fire emissions and Asian dusts tend to elevate the aerosol extinction mainly above the ABL (from 2-3 km up to 6 km) over the Northeastern Pacific, the enhancement of aerosol extinction in the ABL results largely from transport of industrial pollution aerosol (*Bertschi et al.*, 2004; *Price et al.*, 2003; *de Gouw et al.*, 2004). Air masses from Southeast Asia and China are generally observed at higher altitudes than air from Japan and Korea (*de Gouw et al.*, 2004). Tracer model simulations suggest that Asian pollution transported to the west coast of the Northwestern U.S. was largely confined to the lowest 3 km layer, whereas the isentropic trajectory indicates a somewhat higher transport height (*Jaffe et al.*, 1999).

Multi-year measurements from the ground-based micro-pulse lidars in the Korean peninsula (*Kim et al.*, 2007) indicate that, on an annual basis, roughly 2/3 of columnar AOD is contributed by aerosols below 2 km. In comparison to summer and fall measurements, aerosol extinction in spring and winter is enhanced above the boundary layer and below about 6 km, which is presumably attributable to seasonal variations of Asian dust storms. Lidar measurements over Japan and Korea during the ACE-Asia give the similar vertical distribution for polluted cases (*Quinn et al.*, 2004). Observations of aerosol profiles from GLAS are also examined for two available periods. **Figure 1** shows profiles of the frequency and average backscatter of GLAS detected aerosol layers during September-November, 2003 (Figure 1b) and February-March, 2004 (Figure 1c) in four coastal regions as illustrated in Figure 1a. While aerosol layers are largely detected in the boundary layer in both periods and all regions, some enhanced layers with backscatter comparable to that in the ABL are indeed detected in the free atmosphere. Such enhancement in the free atmosphere is more pronounced in February-March than in September-November, presumably because of more frequent dust plumes in late winter

and spring that generally transports at higher altitudes than pollution plumes. Note also that the detected aerosol in the ABL could also be partially attributable to the maritime aerosol. It is thus difficult to characterize pollution aerosol from only GLAS aerosol measurements.

To be consistent with the observations and models discussed above, we assume that the aerosol optical depth in each of three layers, namely 0-1 km, 1-2 km, and above 2 km (denoted respectively as L1, L2, and L3), accounts for one third of the columnar pollution AOD. That is, 67% of pollution AOD is assume to reside in the lowest 2 km layer (approximately, ABL) and 33% above the ABL. This is a simplified representation for the complicated layering structure of aerosols in reality. Seasonal, interannual, and geographical variations of pollution transport heights are not considered in this study because of the paucity of available observations. Therefore seasonal, interannual, and geographical variations of pollution fluxes discussed in this study result from a combination of the MODIS observed variations of column aerosol mass loading and variations of wind and humidity profiles.

3.3. Calculating pollution aerosol mass loading and flux

Pollution aerosol optical depth (τ_p) derived above is for hydrated pollution aerosol in ambient conditions measured by MODIS. In each layer (l), τ_p can be converted to dry mass loading M_p as follows:

$$M_p(l) = \frac{\tau_p(l)}{f[RH(l)]MEE_p} \quad (2)$$

where MEE_p ($m^2 g^{-1}$) is dry mass extinction efficiency of pollution aerosol; and $f(RH)$ is a function that accounts for an increase of aerosol extinction with increasing relative humidity (RH), depending on chemical composition with monotonic (smoothly varying) or deliquescent

(step change) growth (*Quinn et al.*, 2005; *Carrico et al.*, 2003). For MEE_p we use a value of $4 \text{ m}^2\text{g}^{-1}$ based on measurements during ACE-Asia campaign for continental outflows with weak dust influence and at low RH (*Bates et al.*, 2006 and references therein). For $f(\text{RH})$ we use an empirical function that is an average of the monotonic and deliquescent growth curves observed during ACE-Asia for pollution aerosol (*Carrico et al.*, 2003). Measurements of RH from the Atmospheric Infrared Sounder (AIRS) (*Aumann et al.*, 2003) are used as input to quantify the hygroscopic growth of aerosols and convert the measured optical depth in ambient conditions to the (dry) mass loading. To comply with vertical resolution of AIRS datasets, we use RH at 925, 850, and 700 hPa to derive pollution mass loading in L1, L2, and L3 layer, respectively.

The daily mass loading (gm^{-2}) for pollution aerosol derived at individual $1^\circ \times 1^\circ$ grids is aggregated to the monthly average (M_p) over the flux calculation boxes. Then monthly average zonal winds at 925, 850, and 700 hPa from the Goddard Earth Observing System Data Assimilation System (*GEOS DAS*) with current version of GEOS-4 are used to calculate pollution fluxes (gs^{-1}) in L1, L2, and L3 layer, respectively. For a segment with a length of L (m) that is parallel to longitudes, we use east-west wind component U to calculate the columnar fluxes as follows:

$$F_p = \sum_{l=1}^3 M_p(l) U(l) * L \quad (3)$$

The calculated monthly fluxes are aggregated into seasons and 10° latitude sections for analysis in the next section.

4. Results and Discussion

We applied the method discussed in Section 3 to MODIS/Terra Level 3 daily $1^\circ \times 1^\circ$ data from 2002-2005 to estimate fluxes of pollution aerosol exported from East Asia to West Pacific (WP) and imported to the West Coast of North America over East Pacific (EP). The flux estimation was done across meridional planes with a width of 10° in longitude, as illustrated in **Figure 2**. Over West Pacific, the meridional plane is centered at 130°E south of 40°N and at 140°E north of 40°N . On the other side of Pacific, the meridional plane is centered at 130°W . We integrate the calculated flux from 20°N to 60°N over WP to represent the East Asia outflow. Given that the transpacific transport usually shifts poleward after leaving East Asia and the transport in subtropical (20° - 30°N) EP is predominately controlled by easterlies, fluxes are integrated from 30°N to 60°N over EP to represent North America inflow. In the following, seasonal and latitudinal variations of MODIS-estimated export and import fluxes are discussed and compared with GOCART and GMI simulations in Section 4.1. Interannual variability of pollution fluxes are examined for 2002-2005 period in Section 4.2. In Section 4.3, uncertainties associated with flux calculations are estimated and a potential of reducing the uncertainty is discussed in terms of integrating A-Train observations.

4.1. Seasonal and latitudinal variations of pollution fluxes

Since most field experiments and promoted modeling studies of aerosols have been focusing on late winter and spring (e.g., *Tan et al.*, 2002; *Chin et al.*, 2004; *Heald et al.*, 2006; *Hadley et al.*, 2007), seasonal variations of transpacific transport of pollution aerosol are not well understood. **Figure 3** shows the MODIS-estimated seasonal pollution fluxes for 2004. Clearly, both East Asia outflow and North America inflow peaked in spring, with the flux of 6.8 and 1.7 Tg/season, respectively. The outflow and inflow are weakest but not negligible in summer, with

a magnitude of about 1/3 of the springtime maximum and 10% of the annual flux. Winter and autumn appear to be transitional seasons when pollution fluxes are in between the springtime maximum and the summertime minimum. Over the western Pacific, the wintertime pollution flux is about 30% higher than that in autumn. Such seasonal variation for pollution aerosol fluxes is similar to simulations of carbon monoxide (*Liu et al.*, 2003).

The observed seasonal variations of pollution aerosol fluxes are determined by a combination of meteorological conditions, emissions, chemistry and removal processes. In East Asia, emissions of pollution aerosols and their precursors have some seasonal variations. In winter the use of coal in northern China is more than those in other seasons because of heating demands in winter. On the other hand, photochemical production of sulfate aerosol from the combustion generated SO₂ is slower in the winter because of lower oxidant levels and slower reaction rates. Biomass burning shows strong seasonal variations; In South and Southeast Asia, the burning occurs mainly in spring, while in high latitude Eurasian region, the burning peaks in spring and summer (e.g., *Giglio et al.*, 2006; *Bian et al.*, 2007). Presumably the Asian monsoon system would play a much larger role in regulating the seasonality of trans-Pacific transport, as suggested by a number of previous studies. The WCBs are stronger and occur more frequently in winter and spring (*Eckhardt et al.*, 2004) and the springtime dry convection can also be an important lifting mechanism over China (*Dickerson et al.*, 2007). This in combination with strongest mid-latitude westerlies results in the strongest pollutant transport in spring. The weakest pollutant transport in summer is a result of an offset of the westward transport by the eastward transport, particularly south of 30°N and the largest aerosol removal associated with summer monsoon circulation (*Holzer et al.*, 2005).

On an annual basis, the MODIS-estimated pollution outflow and inflow is 16.0 and 4.2 Tg/year, respectively (**Table 1**). This suggests that about a quarter of East Asia outflow of pollution aerosol can reach the west coast of North America, if there is no additional contribution of pollution aerosol from elsewhere during trans-Pacific transport. While this would be a reasonable assumption on an annual basis, the assumption may overestimate Asia's contribution to North America in summer season and certain years because of potential contributions of biomass burning smoke from Alaska. The smoke from Mexico and Central America in spring is unlikely to contribute significantly to the calculated North America inflow over 30°-60°N. By comparison, GOCART and GMI simulations give an outflow of about 10% smaller than the MODIS estimate. On the other hand, both GOCART and GMI give 4.8 Tg/year for the North America inflow, i.e., 15% larger than the MODIS-based estimate. The model simulations suggest that about one third of East Asia outflow can reach the west coast of North America, which is somewhat larger than the MODIS-based estimate. **Figure 4** shows that on a seasonal basis satellite estimated and model simulated outflow and inflow fluxes are still in reasonably good agreement. The largest satellite-model differences occurred in spring for the East Asia outflow (as large as about 30% or 2 Tg) and in summer for the North America inflow (as large as 70% or 0.5 Tg).

Meridional variations of annual pollution aerosol fluxes are compared between satellite estimate and model simulations, as shown in **Figure 5**. For East Asia outflow, MODIS estimated the maximum flux of 7.6 Tg/year in the 30°-40°N segment, followed by 5.7 Tg/year in the 40°-50°N segment. The flux integrated over the 30°-50°N segment accounts for about 80% of the

total East Asia outflow. A remaining 20% of the outflow is nearly evenly distributed in the 20°-30°N and 50°-60°N segments. For comparison, GOCART simulation seems to shift the pollution outflow northward. The maximum outflow occurs in the 40°-50°N segment, larger than the corresponding MODIS estimate by about 20%. On the other hand, the GOCART simulation of outflow is larger than the MODIS estimate by 75% in the 50°-60°N segment but smaller by 60% in the 30°-40°N segment. GMI simulations give that major outflow path is situated in the 30°-50°N segment, consistent with the MODIS estimate. However the magnitude of flux is about 25% lower than the MODIS estimates. On the other hand, the GMI flux of 2.9 Tg/year in the 20°-30°N segment is a factor of more than 2 larger than the MODIS estimate.

For the North America inflow, MODIS estimates the highest flux of 1.8 Tg/year in the 40°-50°N segment, followed by 1.4 Tg/year in the 50°-60°N segment and 1.1 Tg/year in the 30°-40°N segment. This suggests a general poleward shift of pollution plume during the trans-Pacific transport. In the subtropical segment (20°-30°N), the pollution flux is negative, suggesting a westward transport of pollution from North America. GOCART fluxes are higher in the 40°-60°N segments but lower in the 20°-40°N segments than the MODIS estimates. Similar to that over West Pacific, GMI seems to shift the inflow toward lower latitudes, with its flux in the 30°-40°N segment substantially larger than both MODIS and GOCART fluxes. In the 20°-30°N subtropical segment the GMI flux has a small but positive value, suggesting a weak eastward transport. This difference is likely to result from different transport heights in conjunction with the shift of westerly boundary layer winds to easterlies in the free troposphere.

The above analysis suggests that in 2004 about 16Tg pollution aerosol was exported from East Asia to the northwestern Pacific Ocean through mainly the 30°-50°N segment. The pollution flux was largest in spring and smallest in summer. The summer flux was about 30% of the springtime flux. About one quarter of the pollution export from Asia may have reached the west coast of North America in that year. This substantial trans-Pacific transport of pollution is arguably mostly originating from Asia, with some contributions from Europe, Africa, and elsewhere depending on season and location (*Newell and Evans, 2000; Bey et al., 2001*). The satellite-based estimates agree with GOCART and GMI simulations remarkably well over broad meridional segments and on an annual basis. The comparisons also show reasonably good agreements over 10° meridional segments and on a seasonal basis. In comparison to MODIS estimates, GOCART simulations generally shift the pollution transport northward while GMI simulations do otherwise.

4.2. Interannual variability of pollution fluxes

While some interannual variations or trends of industrial emissions in East Asia have been suggested (e.g., *Carmichael et al., 2002; Richter et al., 2006*), biomass burning emissions that are controlled by meteorological and biospheric conditions as well as human activities are likely to show much larger interannual variations (*Duncan et al., 2003*). On the other hand, the interannual variability of Asian outflow resulting from large-scale variations in atmospheric circulation is the lowest, in comparison to other regions (*Liu et al., 2005*). It is thus expected that pollution fluxes exported from East Asia and imported to North America would show some interannual variations resulting mainly from variations in biomass burning smoke from Eurasia.

We have applied the method to MODIS/Terra data for other years and derived the four-year climatology (2002-2005) of pollution fluxes exported to the West Pacific and imported to the East Pacific. **Figure 6** shows interannual variations of pollution aerosol optical depth in MAM and JJA. In MAM and JJA, 2003, pollution aerosol optical depth in the high-latitude North Pacific (40° - 60° N) was elevated to a level that is more than a factor of 2 larger than in other years. This is corroborated with elevated TOMS absorbing aerosol index that suggests intense biomass burning smoke (not shown). In 2003, biomass fires burned a large area of south and east Russia, starting from May through the summer (*Goldammer et al.*, 2004; *Wotawa et al.*, 2006). Both the fire pixel count and burned area are a factor of 3-4 larger than that in 2004 (*Wotawa et al.*, 2006). In fact the burned area in 2003 was the largest in a decade. The pollution aerosol optical depth in high-latitudes (40° - 60° N) is about a factor of 3 larger in 2003 than 2004. On the other hand, boreal forest fires in Alaska and Canada in summer were more intense in 2004 than in 2003. The burned area and fire counts were a factor of 2-3 larger in 2004 than in 2003. Although such boreal fires mainly influence the North America continent in the eastern and southeastern U.S. (*Wotawa and Trainer*, 2000), there is a discernable influence on the surrounding northeastern Pacific, as shown in the MODIS pollution aerosol map. In the mid-latitude West Pacific, pollution largely from China influenced the region to the greatest extent in MAM and JJA, 2005. In the tropical East Pacific, the spring-time biomass burning smoke from Central America was weakest in 2004.

Table 2 compares annual pollution fluxes of East Asia outflow and North America inflow from 2002 to 2005. The annual pollution flux varies by as much as 25% for the outflow and 45% for the inflow. Both the outflow and inflow were strongest in 2003, because of record

intense biomass burning emissions from spring to summer in Eurasia. About 20 Tg pollution aerosol was transported from East Asia to the northwestern Pacific, of which 28% reached the west coast of North America. This is followed by 2005 when 18.6 Tg pollution aerosol was exported from East Asia and 4.1 Tg was imported to North America. As discussed earlier, the pollution AOD in 2005 was higher than that in other years south of 40°N. While the pollution outflow was more or less of 16 Tg in 2002 and 2004, the inflow in 2002 is 12% smaller than that in 2004. The four-year average pollution outflow is 17.7 Tg/year, of which about 25% (or 4.4 Tg/year) can reach the west coast of North America.

The interannual variation of pollution fluxes depends strongly on season and latitude, as shown in **Figure 7** for the East Asia outflow. In the 30°-40°N segment, the springtime pollution flux in 2005 is about 180% higher than that in 2003, with that in 2002 and 2004 in between. The 2005 summertime pollution flux is a factor of 2-3 higher than that of 2002 and 2004, and 30% higher than that of 2003. In latitudes north of 40°N, the springtime pollution flux in 2003 is about a factor of 2 in 40°-50°N and a factor of more than 3 higher in 50°-60°N than that in other years, due to record intense boreal forest fires.

4.3. Estimated uncertainties of pollution fluxes

The above-estimated pollution fluxes will inevitably be subject to uncertainties due to assumptions in implementing the algorithm. Major sources of uncertainties contributing to the pollution flux estimation integrated over latitudes and on annual average are listed in **Table 3** and discussed as follows.

One of first uncertainties would result from the MODIS-based estimate of anthropogenic aerosol optical depth. Kaufman et al. (2005b) estimated that the uncertainty in the derived anthropogenic AOD is 30%. Here we assume an uncertainty of 50% to reflect potentially larger uncertainty on regional scales. In this study seasonal and geographical variations of maritime fine-mode fraction have been accounted for. However, a robust validation or evaluation is not possible because of lack of observations of anthropogenic aerosol optical depth.

The mass scattering efficiencies determined by aircraft and ship measurements are generally consistent (*Clarke et al.*, 2002) and their uncertainty comes from measurements of aerosol mass and scattering, which is estimated to be about 25% (*Quinn et al.*, 2004). The uncertainty for the single-scattering albedo is estimated to be 10%. These result in an estimate of overall uncertainty for the mass extinction efficiency to be 30%. For the fractional change in aerosol scattering as a function of RH, measurements from two nephelometers agreed within 10% (*Carrico et al.*, 2003). For pollution aerosol, the hysteresis factor, a ratio of scattering coefficient on the monotonic to deliquescent curves of the hysteresis loop, is estimated to be 1.3 at RH = 60% (*Carrico et al.*, 2003). The use of average of monotonic and deliquescent growth curve would introduce an error of 15%. Therefore we estimate an overall uncertainty of 20% for $f(\text{RH})$.

Polar-orbiting satellite sensors can only observe aerosols once daily and a combined use of the same MODIS sensor onboard the twin EOS satellites Terra (10:30AM local time) and Aqua (1:30PM local time) allows twice daily observations of aerosols in a region. Therefore a fundamental question to be addressed is: to what degree can flux estimates from MODIS/Terra

and/or MODIS/Aqua represent the diurnal average flux in all-sky conditions? While MODIS Terra and Aqua measurements on average well represent the daily aerosol optical depth on a global scale (*Kaufman et al.*, 2000, 2005c) and in East Asia (*Smirnov et al.*, 2002), it is not clear if MODIS-based estimates of pollution fluxes are a good representation of the daily average. Here we use GOCART model simulations to examine this issue by extracting the model calculated pollution fluxes around Terra/Aqua overpass times. It is found that the GOCART calculated fluxes at the Terra/Aqua overpass times generally differ from the GOCART day and night averages by about 10%. These results give us some confidence that aerosol measurements from MODIS-Terra and/or MODIS-Aqua, can be used to quantify daily flux rates with a relatively small and definable uncertainty.

The determination of transport heights of pollution aerosol plume has been used for dual purposes, i.e., selecting representative RH to account for humidification growth of aerosol and selecting representative wind speed for calculating mass fluxes. In this study, we have partitioned pollution AOT into 3 vertical segments of 0-1, 1-2, and above 2 km with equal fractions; a higher transport height (i.e., a larger fraction at higher altitudes) would result in larger aerosol mass flux because of stronger wind speed and drier air mass (hence greater aerosol mass for a given optical depth) at high altitudes in general. The sensitivity of pollution mass flux to the transport height should vary with region, depending on patterns of wind shear and atmospheric humidity. To examine such sensitivity, we assume pollution aerosol is evenly distributed in four vertical layers up to an altitude of 5 km and wind and humidity at 925, 850, 700, and 600 hpa are used to calculate aerosol mass loading and fluxes in individual layers respectively. In comparison to the original assumption of vertical distribution, the pollution flux

increases by 20-80%, depending on season and latitude. By integrating over all latitudes, the East Asia outflow and North America inflow increases by 40-60% and 30-40%, respectively.

The overall uncertainty in the calculated annual pollution aerosol fluxes for East Asia outflow and North America inflow is estimated as ~80%, assuming that the individual uncertainties are independent. Note that the estimated pollution fluxes may have larger uncertainties in some sub-regions and seasons. While biomass burning smoke can come from both natural and manmade fires, it remains challenging to make an unambiguous distinction between them. Our assumption that all biomass burning aerosols are pollution would have biased our estimates of pollution fluxes in high latitudes in spring and summer where Eurasia boreal forest fires usually occur. On the other hand, the determination of transport heights and hygroscopic properties of pollution aerosol in this study is largely based on field experiments that have focused on industrial/urban pollution. This may have underestimated the flux of smoke aerosol because smoke aerosol can be raised to higher altitudes where wind speed is generally stronger, humidity is lower, and smoke aerosol usually has lower hygroscopicity (i.e., a larger mass loading for smoke than for industrial pollution for a given AOD).

In this study we use the monthly mean dry aerosol mass derived from satellite retrievals in cloud-free conditions to calculate transport flux of pollution aerosol. Inherent in these estimates of pollutant transport is the assumption that the dry mass transport over the course of a month is the same, whether or not there are frontal cloud systems in the region. Frontal cloud systems extend over wide areas, preventing aerosol retrievals in those areas. These are not the same as broken cloud fields where satellite retrievals of aerosols are possible in the holes

between clouds. For example, because aerosol transport is associated with the WCB, and the WCB is associated with rising air in warm fronts that also consist of wide spread cloudiness, MODIS may miss important transport events every month. If dry aerosol mass in these events is significantly different from the monthly mean observed in cloud-free conditions, the estimated pollutant flux may have large uncertainties or biases. It has been suggested that a variety of compensating processes in and nearby clouds control the formation and removal of aerosols and hence determine their loading. While clouds can effectively remove aerosols from atmosphere through scavenging and rainout, they can also generate aerosol (e.g., in-cloud aqueous production of sulfate). We do not know how to quantify the differences in dry aerosol mass between cloud-free conditions and frontal cloud systems, and whether an overall bias exists in the estimated pollution fluxes. However, we acknowledge that the 80% uncertainty calculated from the known uncertainties may be a conservative number.

A reduction of uncertainty in the estimated pollution aerosol fluxes can be achieved through integrating measurements from other A-Train sensors flying in formation with MODIS. For example, the new capabilities of passive remote sensing such as MODIS should be further enhanced by emerging lidar measurements of aerosol vertical distributions made by CALIPSO (*Winker et al.*, 2003). GLAS data with limited temporal coverage has been used in this study to help determine aerosol transport heights. However, the availability of only 532 nm GLAS data makes it difficult to separate pollution aerosol from dust. Since June 2006, CALIPSO has been sensing the vertical distributions of aerosol backscatter and extinction in continuous mode, enhancing the temporal coverage of data. Furthermore the use of the two wavelengths and the depolarization capability at 532 nm of CALIOP can be used to better categorize aerosol types. In

the future, fully calibrated and validated CALIPSO aerosol extinction/backscatter profile data can be used to achieve a better quantification of aerosol intercontinental transport, in terms of seasonal, interannual, and geographical variations. A retrieval of fine and coarse-mode separated aerosol extinction profile with a fusion of CALIOP and MODIS/POLDER measurements (Kaufman *et al.*, 2003a,b) would make a reliable estimate of vertical distributions of aerosol mass flux possible.

The Polarization and Directionality of the Earth's Reflectance (POLDER) on the Polarization and Anisotropy of Reflectance for Atmospheric Science coupled with Observations from a Lidar (PARASOL) is measuring directional and polarized radiance in nine spectral channels from 443 nm to 1020 nm. In addition to retrievals of AOD and size parameters, the analysis of the polarized angular scattered radiance provides additional information on the shape of particles (e.g., spherical vs non-spherical) (Herman *et al.*, 2005). This complements the MODIS spectral measurements which are not sensitive to particle shape and can be used to separate non-spherical dust from spherical maritime aerosols. POLDER and MODIS show an excellent agreement when particles are spherical (G  r  rd *et al.*, 2005). Therefore POLDER spherical coarse-mode aerosol optical depth over ocean can be used to separate dust from maritime aerosol in coarse-mode. The Ozone Monitoring Instrument (OMI) onboard Aura since June 2004 and its predecessor, the Total Ozone Mapping Spectrometer (TOMS) can separate UV-absorbing mineral dust or biomass burning smoke from scattering sulfate aerosol (Herman *et al.*, 1997). Such information could be used to determine if the plume is dominated by industrial pollution or smoke and hence to make a better characterization of aerosol growth with relative humidity.

5. Summary and Conclusion

By taking advantage of MODIS high accuracy measurements of aerosol optical depth and fine-mode fraction over oceans, we have developed an observation-based approach to estimate the intercontinental transport of pollution aerosol over the North Pacific. For this method, measurements from other satellite sensors, such as AIRS and GLAS, and from field campaigns have also been used. We estimated that for 2002-2005 average, about 18 Tg/year of pollution aerosol is exported from Asia, of which about 25% arrives in the west coast of North America. The export and import pollution fluxes are largest in spring and smallest in summer, with the springtime and summertime flux accounting for about 40% and 10% of the annual integrated flux, respectively. For the period we have examined the strongest pollution export occurred in 2003, due largely to record intense boreal forest fires in Eurasia. The overall uncertainty of pollution fluxes is estimated at 80%, due to uncertainties in separating pollution aerosol from dust and maritime aerosol, in deriving the aerosol mass loading and determining the transport heights of the pollution plume. A reduction of uncertainty can be achieved with a better characterization of pollution aerosol through integrating the PARASOL measurements of spherical and non-spherical aerosols, CALIPSO measurements of vertical distributions of pollution aerosol, and OMI aerosol indices. Simulations by the Goddard Ozone Chemistry Aerosol Radiation and Transport (GOCART) and Global Modeling Initiative (GMI) models agree quite well with the satellite-based estimates of annual and latitude-integrated fluxes, with larger model-satellite differences in latitudinal variations of fluxes. The satellite-based approach developed in this study can be adapted and extended to study the intercontinental transport of pollution in other regions.

While this study shows that a substantial amount of pollution aerosol from Asia can be transported to North America, its implications for climate change and air quality have not been assessed. Both modeling studies with reliable representations of major transport mechanisms at a wide range of scales and analyses of CALIPSO aerosol vertical distributions are necessary for gaining insights of downward transport of the height-elevated pollution to the atmospheric boundary layer and of potential influences of Asian pollution aerosol on surface air quality in North America. Further research is needed to assess how the intercontinental transport of pollution aerosol can influence the weather and climate over the North Pacific and how the resulting changes in atmospheric circulations would feedback on the transpacific transport of pollution and dust. Long-term trends in the transpacific pollution transport and its environmental impacts would be detected with an accumulation of satellite data in the future.

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References:

- ✓ Akimoto, H., T. Ohara, J. Kurokawa, and N. Horii (2006), Verification of energy consumption in China during 1996-2003 by using satellite observational data, *Atmos. Environ.*, **40**, 7663-7667.
- ✓ Al-Saadi, J., et al. (2005), Improving national air quality forecasts with satellite aerosol observations, *Bull. Am. Met. Soc.*, **86**(9), 1249-1261.
- ✓ Anderson, T.L., et al., (2005a), A-Train strategy for quantifying direct climate forcing by aerosols, *Bull. Amer. Meteorolo. Soc.*, **86**(12), 1795-1809.
- Anderson, T.L., et al. (2005b): Testing the MODIS satellite retrieval of aerosol fine-mode fraction, *J. Geophys. Res.*, **110**, D18204, doi:10.1029/2005JD005978.
- ✓ Andreae, M.O., et al. (1988), Vertical distribution of dimethylsulfide, sulfur dioxide, aerosol ions, and radon over the North-East Pacific Ocean, *J. Atmos. Chem.*, **6**, 149-173.
- ✓ Aumann, H., et al. (2003), AIRS/AMSU/HSB on the Aqua mission: Design, science objectives, data products, and processing system, *IEEE Trans. Geoscience & Remote Sensing*, **41**(2), 253-264.
- ✓ Baheini, R., et al. (2003), Aircraft-based aerosol size and composition measurements during ACE-Asia using an Aerodyne aerosol mass spectrometer, *J. Geophys. Res.*, **108** (D23), 8645, doi:10.1029/2002JD003226.
- ✓ Bates, T.S., et al. (2006), Aerosol direct radiative effects over the northwest Atlantic, northwest Pacific, and North Indian Oceans: estimates based on in-situ chemical and optical measurements and chemical transport modeling, *Atmos. Chem. Phys.*, **6**, 1657-1732.
- ✓ Bates, T.S., et al. (2001), Regional physical and chemical properties of the marine boundary layer aerosol across the Atlantic during Aerosols99: An overview, *J. Geophys. Res.*, **106**(D18), 20767-20782.
- ✓ Bellouin N, Boucher O, Haywood J, and M.S. Reddy (2005), Global estimate of aerosol direct radiative forcing from satellite measurements, *Nature*, **438** (7071): 1138-1141, doi:10.1038/nature04348.
- ✓ Bertschi, I.T., D.A. Jaffe, L. Jaegle, H.U. Price, and J.B. Dennison (2004), PHOBEA/ITCT2002 airborne observations of transpacific transport of ozone, CO, volatile organic compounds, and aerosols to the northeast Pacific: Impacts of Asian anthropogenic and Siberian boreal fire emissions, *J. Geophys. Res.*, **109**, D23S12, doi:10.1029/2003JD004328.
- ✓ Bey, I., D.J. Jacob, J.A. Logan, and R.M. Yantosca (2001), Asian chemical outflow to the Pacific in spring: Origins, pathways, and budgets, *J. Geophys. Res.*, **106**, 23097-23114.
- ✓ Bian, H., M. Chin, R. Kawa, B. Duncan, A. Arellano Jr., and R. Kasibhatla (2007), Uncertainty of global CO simulations constraint by biomass burning emissions. Submitted to *J. Geophys. Res.*, .
- ✓ Biscaye, P., F.E. Grousset, A.M. Svensson, and A. Bory (2000), Eurasian air pollution reaches eastern North America, *Science*, **290**, 2258-2259, doi:10.1126/science.290.5500.2258.

- ✓ Bond, T., D. Streets, K. Yarber, S. Nelson, J. Woo, and Z. Klimont (2004), A technology-based inventory of black and organic carbon emissions from combustion, *J. Geophys. Res.*, *109*, D14203, doi:10.1029/2003JD003697.
- ✓ Carmichael, G., D. Streets, G. Calori, M. Amann, M.Z. Jacobson, J. Hansen, and H. Ueda (2002), Changing trends in sulfur emissions in Asia: Implications for acid deposition, air pollution, and climate, *Environ. Sci. Technol.*, *36*, 4707-4713.
- ✓ Carrico, C.M., P. Kus, M.J. Rood, P.K. Quinn, and T.S. Bates (2003), Mixtures of pollution, dust, sea salt, and volcanic aerosol during ACE-Asia: Radiative properties as a function of relative humidity, *J. Geophys. Res.*, *108*(D23), 8650, doi:10.1029/2003JD003405.
- ✓ Chameides, W. L., H. Yu, S.C. Liu, et al. (1999): A case study of the effects of atmospheric aerosols and regional haze on agriculture: An opportunity to enhance crop yields in China through emission controls? *Proc. Natl. Acad. Sci.*, *96*(24), 13626-13633.
- ✓ Chin, M., R. B. Rood, S.-J. Lin, J. F. Muller, and A. M. Thomsson (2000a), Atmospheric sulfur cycle in the global model GOCART: Model description and global properties, *J. Geophys. Res.*, *105*, 24,671-24,687.
- ✓ Chin, M., D. Savoie, B. J. Huebert, et al. (2000b), Atmospheric sulfur cycle in the global model GOCART: Comparison with field observations and regional budgets, *J. Geophys. Res.*, *105*, 24,689-24,712.
- ✓ Chin, M., P. Ginoux, S. Kinne, et al. (2002), Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sunphotometer measurements, *J. Atmos. Sci.*, *59*, 461-483.
- ✓ Chin, M., D. A. Chu, R. Levy, et al. (2004), Aerosol distribution in the Northern Hemisphere during ACE-Asia: Results from global model, satellite observations, and Sun photometer measurements. *J. Geophys. Res.*, *109* (D23), doi: 10.1029/2004GL02014.
- ✓ Chin, M., T. Diehl, P. Ginoux, and W. Malm (2007), Intercontinental transport of pollution and dust aerosols: implications for regional air quality, *Atmos. Chem. Phys. Discuss.*, *7*, 9013-9051.
- ✓ Christopher, S.A., et al. (2006), Satellite-based assessment of top of atmosphere anthropogenic aerosol radiative forcing over cloud-free oceans, *Geophys. Res. Lett.*, *33*, L15816, doi:10.1029/2005GL025535.
- ✓ Chu, D. A., L. A. Remer, Y. J. Kaufman, et al. (2005), Evaluation of aerosol properties over ocean from Moderate Resolution Imaging Spectroradiometer (MODIS) during ACE-Asia. *J. Geophys. Res.*, *110* (D07308), doi: 10.1029/2004JD005208.
- ✓ Chung, Y.S. (1986), Air pollution detection by satellites: The transport and deposition of air pollutants over oceans, *Atmos. Environ.*, *20*, 617-630.

- ✓ Clarke, A.D., et al. (2002), INDOEX aerosol : A comparison and summary of chemical, microphysical, and optical properties observed from land, ship, and aircraft, *J. Geophys. Res.*, 107(D19), 8033, doi:10.1029/2001JD000572.
- ✓ de Gouw, J.A., et al. (2004), Chemical composition of air masses transported from Asia to the U.S. West Coast during ITCT 2K2: Fossil fuel combustion versus biomass-burning signatures, *J. Geophys. Res.*, 109, D23S20, doi:10.1029/2003JD004202.
- ✓ Dibb, J.E., et al., (2003), Aerosol chemical composition in Asian continental outflow during the TRACE-P campaign: Comparison with PEM-West B, *J. Geophys. Res.*, 108(D21), 8815, doi:10.1029/2002JD003111.
- ✓ Dickerson, R.R., et al. (2007), Aircraft observations of dust and pollutants over NE China: Insight into the meteorological mechanisms of long-range transport, *J. Geophys. Res.*, *EAS-AIRE Special Issue*, in press.
- ✓ Duce, R.A., C.K. Unni, and B.J. Ray (1980), Long-range atmospheric transport of soil dust from Asia to the tropical North Pacific: Temporal variability, *Science*, 209(26), 1522-1524.
- ✓ Duncan, B., R. Martin, A. Staudt, R. Yevich and J. Logan (2003), Interannual and Seasonal Variability of Biomass Burning Emissions Constrained by Satellite Observations, *J. Geophys. Res.*, 108, doi:10.1029/2002JD002378.
- ✓ Eckhardt, S., et al. (2004), A 15-year climatology of warm conveyor belts, *J. Climate*, 17, 218-237.
- ✓ Engel-Cox, J.A., C.H. Holloman, B.W. Coutant and R.M. Hoff (2004), Qualitative and quantitative evaluation of MODIS satellite sensor data for regional and urban scale air quality, *Atmos. Environ.*, 38, 2495-2509.
- ✓ Fairlie, T.D., D.J. Jacob, R.J. Park (2007), The impact of transpacific transport of mineral dust in the United States, *Atmos. Environ.*, 41, 1251-1266.
- ✓ Fraser, R.S. (1976), Satellite measurement of mass of Sahara dust in the atmosphere, *Appl. Opt.*, 15, 2471-2479.
- ✓ Fraser, R.S., Y.J. Kaufman, and R.L. Mahoney (1984), Satellite measurements of aerosol mass and transport, *Atmos. Environ.*, 18, 2577-2584.
- ✓ Gérard, B., et al. (2005), Comparisons between POLDER2 and MODIS/Terra aerosol retrievals over ocean, *J. Geophys. Res.*, 110, D24211, doi:10.1029/2005JD006218.
- ✓ Giglio, L., G.R. van der Werf, J.T. Randerson, G.J. Collatz, and P. Kasibhatla (2006), Global estimation of burned area using MODIS active fire observations, *Atmos. Chem. Phys.*, 6, 957-974.
- ✓ Ginoux, P., M. Chin, I. Tegen, J. Prospero, B. Holben, O. Dubovik, and S.-J. Lin (2001), Sources and distributions of dust aerosols simulated with the GOCART model, *J. Geophys. Res.*, 106, 20225-20273.

- ✓ Ginoux, P., J. Prospero, O. Torres, and M. Chin (2004): Long-term simulation of dust distribution with the GOCART model: Correlation with the North Atlantic Oscillation, *Environ. Modeling and Software*, 19, 113-128.
- ✓ Goldammer, J.G., A. Sukhinin, and I. Csiszar (2004), The current fire situation in the Russian Federation: Implications for enhancing international and regional cooperation in the UN framework and the global programs on fire monitoring and assessment, *Int. Forest Fire News*, 29, 89-111.
- ✓ Grousset, F., P. Ginoux, A. Bory, and P.E. Biscaye (2003), Case study of a Chinese dust plume reaching the French Alps, *Geophys. Res. Lett.*, 30(6), 1277, doi:10.1029/2002GL016833.
- ✓ Hadley, O.L., V. Ramanathan, G.R. Carmichael, Y. Tang, C.E. Corrigan, G.C. Roberts, and G.S. Mauger (2007), Trans-Pacific transport of black carbon and fine aerosols ($D < 2.5 \mu\text{m}$) into North America, *J. Geophys. Res.*, 112, D05309, doi:10.1029/2006JD007632.
- ✓ Hansen, J., and L. Nazarenko (2004), Soot climate forcing via snow and ice albedos, *Proc. Natl. Acad. Sci. USA*, 101(2), 423-428.
- ✓ Heald, C.L., et al. (2006), Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States, *J. Geophys. Res.*, 111, D14310, doi:10.1029/2005JD006847.
- ✓ Herman, J., P. Bhartia, O. Torres, C. Hsu, C. Seftor, and E. Celarier (1997), Global distribution of UV-absorbing aerosols from Nimbus-7/TOMS data. *J. Geophys. Res.* 102, 16911-16922.
- ✓ Herman, M., J.L. Deuzé, A. Marchand, B. Roger, and P. Lallart (2005), Aerosol remote sensing from POLDER/ADEOS over the ocean: Improved retrieval using a nonspherical particle model, *J. Geophys. Res.*, 110, D10S02.
- ✓ Higurashi, A., and T. Nakajima (2002), Detection of aerosol types over the East China sea near Japan from four-channel satellite data, *Geophys. Res. Lett.*, 29(17), 1836, doi:10.1029/2002GL015357.
- ✓ Hoell, J. M., D.D. Davis, S.C. Liu, R. Newell, M. Shipham, H. Akimoto, R.J. McNeal, R.J. Bemdura, and J.W. Drewry (1996), Pacific Exploratory Mission-West A (PEM-WEST A): September-October, 1991, *J. Geophys. Res.*, 101, 1641-1653.
- ✓ Hoell, J. M., D.D. Davis, S.C. Liu, R. Newell, M. Shipham, H. Akimoto, R.J. McNeal, R.J. Bemdura, and J.W. Drewry (1997), The Pacific Exploratory Mission-West Phase B: February-March, 1994, *J. Geophys. Res.*, 102, 28223-28239.
- ✓ Holzer, M., T.M. Hall, and R.B. Stull (2005), Seasonality and weather-driven variability of transpacific transport, *J. Geophys. Res.*, 110, D23103, doi:10.1029/2005JD006261.
- ✓ Holzer, M., and T.M. Hall (2007), Low-level transpacific transport, *J. Geophys. Res.*, 112, D09103, doi:10.1029/2006JD007828.
- ✓ Huang, Y., R.E. Dickinson, and W.L. Chameides (2006), Impact of aerosol indirect effect on surface temperature over East Asia, *Proc. Natl. Acad. Sci.*, 103(12), 4371-4376.

- ✓ Huebert, B.J., T. Bates, P.B. Russell, et al. (2003), An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climatic impacts, *J. Geophys. Res.*, *108*, D23, 8633, doi:10.1029/2003JD003550.
- ✓ Huebert, B.J., T. Bertram, J. Kline, S. Howell, D. Eatough, and B. Blomquist (2004), Measurements of organic and elemental carbon in Asian outflow during ACE-Asia from the NSF/NCAR C-130, *J. Geophys. Res.*, *109*, D19S11, doi:10.1029/2004JD004700.
- ✓ Husar, R.B., et al. (2001), The Asian dust events of April, 1998, *J. Geophys. Res.*, *106*, 18317-18330.
- ✓ Jacob, D.J., et al. (2003), The transport and chemical evolution over the Pacific (TRACE-P) aircraft mission: Design, execution, and first results, *J. Geophys. Res.*, *108*(D20), 9000, doi:10.1029/2002JD003276.
- ✓ Jaffe, D., et al. (1999), Transport of Asian air pollution to North America, *Geophys. Res. Lett.*, *26*, 711-714.
- ✓ Jaffe, D., S. Tamura, and J. Harris (2005), Seasonal cycle and composition of background fine particles along the west coast of the US, *Atmos. Environ.*, *39*, 297-306.
- ✓ Jirak, I.L., and W.R. Cotton (2006), Effect of air pollution on precipitation along the Front Range of the Rocky Mountains, *J. Appl. Meteor. Clim.*, *45*, 236-245.
- ✓ Kaufman, Y.J., I. Koren, L.A. Remer, D. Tanré, P. Ginoux, and S. Fan (2005a), Dust transport and deposition observed from the Terra-Moderate Resolution Imaging Spectroradiometer (MODIS) spacecraft over the Atlantic Ocean, *J. Geophys. Res.*, *110*, D10S12, doi:10.1029/2003JD004436.
- ✓ Kaufman, Y.J., O. Boucher, D. Tanré, M. Chin, L.A. Remer, and T. Takemura (2005b), Aerosol anthropogenic component estimated from satellite data. *Geophys. Res. Lett.* *32*, L17804, doi:10/1029/2005GL023125.
- ✓ Kaufman, Y.J., L.A. Remer, D. Tanré, et al. (2005c), A critical examination of the residual cloud contamination and diurnal sampling effects on MODIS estimates of aerosol over ocean. *IEEE TGRS* *43* (12), 2886-2897.
- ✓ Kaufman, Y. J., I. Koren, L. A. Remer, D. Rosenfeld, and Y. Rudich (2005d), The effect of smoke, dust and pollution aerosol on shallow cloud development over the Atlantic Ocean. *Proc. Natl. Acad. Sci.*, *102* (32), 11207-11212.
- ✓ Kaufman, Y. J., J. M. Haywood, P. V. Hobbs, W. Hart, R. Kleidman, and B. Schmid (2003a), Remote sensing of vertical distributions of smoke aerosol off the coast of Africa. *Geophys. Res. Lett.*, *30*(D16), 1831, doi: 10.1029/2003GL017068.
- ✓ Kaufman, Y. J., D. Tanré, J.-F. León, and J. Pelon (2003b), Retrievals of profiles of fine and coarse aerosols using lidar and radiometric space measurements. *IEEE Trans. Geosci. Remote Sens.*, *41*(8), 1743-1754.

- ✓ Kaufman, Y. J., D. Tanré, and O. Boucher (2002), A satellite view of aerosols in the climate system. Review. *Nature*, 419, 215-223.
- ✓ Kaufman, Y. J., B.N. Holben, D. Tanré, I. Slutsker, A. Smirnov, and T.F. Eck (2000), Will aerosol measurements from Terra and Aqua polar orbiting satellites represent daily aerosol abundance and properties?, *Geophys. Res. Lett.*, 27, 3861-3864.
- ✓ Kaufman, Y. J., D. Tanré, L. Remer, E. Vermote, A. Chu, and B. N. Holben (1997), Operational remote sensing of tropospheric aerosol over land from EOS Moderate Resolution Imaging Spectroradiometer. *J. Geophys. Res.*, 102, 17051-17067.
- ✓ Kim, S., S. Yoon, J. Kim, and S. Kim (2007), Seasonal and monthly variations of columnar aerosol optical properties over East Asia determined from multi-year MODIS, LIDAR, and AERONET Sun/sky radiometer measurements, *Atmos. Environ.*, 41, 1634-1651.
- ✓ King, M. D., Y. J. Kaufman, D. Tanré, and T. Nakajima (1999), Remote sensing of tropospheric aerosols: Past, present, and future, *Bull. Am. Meteorol. Soc.*, 80, 2229-2259.
- ✓ Kleidman, R.G., et al. (2005), Comparison of Moderate Resolution Imaging Spectroradiometer (MODIS) and Aerosol Robotic Network (AERONET) remote-sensing retrievals of aerosol fine mode fraction over ocean, *J. Geophys. Res.*, 110, D22205, doi:10.1029/2005JD005760.
- ✓ Koren, I., Y.J. Kaufman, L.A. Remer, and V. Martin (2004), Measurement of the effect of Amazon smoke on inhibition of cloud formation, *Science*, 303, 1342-1345.
- ✓ León, J.-F., D. Tanré, J. Pelon, Y. J. Kaufman, J. M. Haywood, and B. Chatenet (2003), Profiling of a Saharan dust outbreak based on a synergy between active and passive remote sensing. *J. Geophys. Res.*, 108 (D18), doi: 10.1029/2002 JD002774.
- ✓ Levy, R., L.A. Remer, S. Mattoo, E. Vermote, and Y. Kaufman (2007), Second-generation algorithm for retrieving aerosol properties over land from MODIS spectral reflectance, *J. Geophys. Res.*, 112, D13211, doi:10.1029/2006JD007811.
- ✓ Liang, Q., L. Jaegle, D.A. Jaffe, P. Weiss-Penzias, A. Heckman, and J.A. Snow (2004), Long-range transport of Asian pollution to the northeast Pacific: Seasonal variations and transport pathways of carbon monoxide, *J. Geophys. Res.*, 109, D23S07, doi:10.1029/2003JD004402.
- ✓ Liu, H., D.J. Jacob, I. Bey, R.M. Yantosca, B.N. Duncan, G.W. Sachse (2003), Transport pathways for Asian pollution outflow over the Pacific: Interannual and seasonal variations, *J. Geophys. Res.*, 108(D20), 8786, doi:10.1029/2002JD003102.
- ✓ Liu, J., and J. Diamond (2005), China's environment in a globalizing world, *Nature*, 435, 1179-1186.
- ✓ Liu, J., D.L. Mauzerall, and L.W. Horowitz (2005), Analysis of seasonal and interannual variability in transpacific transport, *J. Geophys. Res.*, 110, D04302, doi:10.1029/2004JD005207.

- ✓ Liu, X., J. Penner, B. Das, D. Bergmann, J. Rodriguez, S. Strahan, M. Wang, and Y. Feng (2007), Uncertainties in global aerosol simulations: Assessment using three meteorological data sets. *J. Geophys. Res.*, *112*, D11212, doi: 10.1029/2006JD008216.
- ✓ Lyons, W.A., J.C. Dooley, Jr., and K.T. Whitby (1978), Satellite detection of long-range pollution transport and sulfate aerosol hazes, *Atmos. Environ.*, *12*, 621-631.
- ✓ McKendry, I.G., et al. (2001), Long-range transport of Asian dust to the Lower Fraser Valley, British Columbia, Canada, *J. Geophys. Res.*, *106*(D16), 18361-18370.
- ✓ Menon, S., J.E. Hansen, L. Nazarenko, and Y. Luo (2002), Climate effects of black carbon aerosols in China and India, *Science*, *297*, 2250-2253, doi:10.1126/science.1075159.
- ✓ Newell, R.E., and M.J. Evans (2000), Seasonal changes in pollutant transport to the North Pacific: the relative importance of Asian and European sources, *Geophys. Res. Lett.*, *27*, 2509-2512.
- ✓ Ohara, T., H. Akimoto, J. Kurokawa, N. Horii, LLK. Yamaji, X. Yan, and T. Hayasaka (2007), An Asian emission inventory of anthropogenic emission sources for the period 1980-2020, *Atmos. Chem. Phys. Discuss.*, *7*, 6843-6902.
- ✓ Parrish, D.D., et al. (2004), Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) and Pacific Exploration of Asian Continental Emission (PEACE) experiments: An overview of the 2002 winter and spring intensives, *J. Geophys. Res.*, *109*, D23S01, doi:10.1029/2004JD004980.
- ✓ Price, H.U., D.A. Jaffe, P.V. Doskey, I. McKendry, and T.L. Anderson (2003), Vertical profiles of O₃, aerosol, CO, NMHCs in the Northeast Pacific during the TRACE-P and ACE-Asia experiments, *J. Geophys. Res.*, *108*(D20), 8799, doi:10.1029/2002JD002930.
- ✓ Qian Y., W. Wang, L Leung, and D. Kaiser (2007), Variability of solar radiation under cloud-free skies in China: The role of aerosols. *Geophys. Res. Lett.* *34*, L12804, doi:10.1029/2006GL028800.
- ✓ Quinn, P.K. (2005), et al., Impact of particulate organic matter on the relative humidity dependence of light scattering: A simplified parameterization, *Geophys. Res. Lett.*, *32*, L22809, doi:10.1029/2005GL024322.
- ✓ Quinn, P.K., et al. (2004), Aerosol optical properties measured on board the Ronald H. Brown during ACE-Asia as a function of aerosol chemical composition and source region, *J. Geophys. Res.*, *109*, D19S01, doi:10.1029/2003JD004010.
- ✓ Remer, L. A., and Y.J. Kaufman (2006), Aerosol direct radiative effect at the top of the atmosphere over cloud free ocean derived from four years of MODIS data. *Atmos. Chem. Phys.* *6*, 237-253.
- ✓ Remer, L.A., Y.J. Kaufman, D. Tanré, et al. (2005), The MODIS aerosol algorithm, products, and validation, *J. Atmos. Sci.*, *62*, 947-973.
- ✓ Remer, L. A., D. Tanré, Y.J. Kaufman, et al. (2002), Validation of MODIS aerosol retrieval over ocean. *Geophys. Res. Lett.*, *29*, doi: 10.1029/2001GL013204.

- ✓ Richter, A., P. Burrows, H. Nues, C. Granier, and U. Niemeijer (2005), Increase in tropospheric nitrogen dioxide over China observed from space, *Nature*, 437, 129-130.
- ✓ Rosenfeld, D., and A. Givati (2006), Evidence of orographic precipitation suppression by air pollution-induced aerosols in the western United States, *J. Appl. Meteor. Clim.*, 45, 893-911.
- ✓ Sassen, K. (2002), Indirect climate forcing over the western US from Asian dust storms, *Geophys. Res. Lett.*, 29(10), 10.1029/2001GL014051.
- ✓ Shirmov, A., B.N. Holben, T.F. Eck, I. Slutsker, B. Chatenet, and R.T. Pinker (2002), Diurnal variability of aerosol optical depth observed at AERONET (Aerosol Robotic Network) sites, *Geophys. Res. Lett.*, 29(23), 2115, doi:10.1029/2002GL016305.
- ✓ Spinhirne, J.D., S.P. Palm, W.D. Hart, D.L. Hlavka, and E.J. Welton (2005), Cloud and aerosol measurements from the GLAS space borne lidar: initial results, *Geophys. Res. Lett.*, 32, L22S03, doi:10.1029/2005GL023507.
- ✓ Stohl, A. (2001), A one-year Lagrangian "climatology" of airstreams in the northern hemisphere troposphere and lowermost stratosphere, *J. Geophys. Res.*, 106, 7263-7279.
- ✓ Stohl, A., S. Eckhardt, C. Forster, P. James, and N. Spichtinger (2002), On the pathways and timescales of intercontinental air pollution transport, *J. Geophys. Res.*, 107(D23), 4684, doi:10.1029/2001JD001396.
- ✓ Stohl, A., C. Forster, H. Huntrieser, et al. (2007), Aircraft measurements over Europe of an air pollution plume from Southeast Asia – aerosol and chemical characterization, *Atmos. Chem. Phys.*, 7, 913-937.
- ✓ Streets, D., and K. Aunan (2005), The importance of China's household sector for black carbon emissions, *Geophys. Res. Lett.*, 32, L12708, doi:10.1029/2005GL022960.
- ✓ Tan, Q., Y. Huang, and W.L. Chameides (2002), Budget and export of anthropogenic SO_x from East Asia during continental outflow conditions, *J. Geophys. Res.*, 107(D13), 4167, 10.1029/2001JD000769.
- ✓ Tanré, D., Y. J. Kaufman, M. Herman, and S. Mattoo (1997), Remote sensing of aerosol properties over oceans using the MODIS/EOS spectral radiances. *J. Geophys. Res.*, 102, 16971-16988.
- ✓ Tanré, D., F.M. Bréon, J.L. Deuzé, M. Herman, P. Goloub, F. Nadal, and A. Marchand (2001), Global observation of anthropogenic aerosols from satellite, *Geophys. Res. Lett.*, 28(24), 4555-4558.
- ✓ VanCuren, R.A., and T.A. Cahill (2002), Asian aerosols in North America: Frequency and concentration of fine dust, *J. Geophys. Res.*, 107, D24, 4804, doi:10.1029/2002JD002204.
- ✓ VanCuren, R.A., S.S. Cliff, K.D. Perry, and M. Jimenez-Cruz (2005), Asian continental aerosol persistence above the marine boundary layer over the eastern North Pacific: Continuous aerosol measurements from International Transport and Chemical Transformation 2002 (ITCT 2K2), *J. Geophys. Res.*, 110, D09S90, doi:10.1029/2004JD004973.

- ✓ Wang, G., K. Kawamura, S. Lee, K. Ho, and J. Cao (2006), Molecular, seasonal, and spatial distributions of organic aerosols from fourteen Chinese cities, *Environ. Sci. Technol.*, **40**, 4619-4625.
- ✓ Wilkening, K. E., L. A. Barrie, and M. Engle (2000), Trans-Pacific air pollution, *Science*, **290**, 65-67.
- ✓ Winker, D.M., J.R. Pelon, and M.P. McCormick (2003), The CALIPSO mission: spaceborne lidar for observations of aerosols and clouds, *Proc. SPIE*, **4893**, 1-11.
- ✓ Wotawa, G., and M. Trainer (2000), The influence of Canadian forest fires on pollutant concentrations in the United States, *Science*, **288**, 324-328.
- ✓ Wotawa, G., L.-E. De Geer, A. Bekcer, R. D'Amours, M. Jean, R. Servranckx, and K. Ungar (2006), Inter- and intra-continental transport of radioactive cesium released by boreal forest fires, *Geophys. Res. Lett.*, **33**, L12806, doi:10.1029/2006GL026206.
- ✓ Xu, X., L. Wang, and T. Niu (1998), Air pollution and its health effects in Beijing, *Ecosystem Health*, **4**, 199-209.
- ✓ Yu, H., R. Dickinson, M. Chin, et al. (2003), Annual cycle of global distributions of aerosol optical depth from integration of MODIS retrievals and GOCART model simulations, *J. Geophys. Res.*, **108**, 4128, doi:10.1029/2002JD02717.
- ✓ Yu, H., R.E. Dickinson, M. Chin, et al. (2004), The direct radiative effect of aerosols as determined from a combination of MODIS retrievals and GOCART simulations, *J. Geophys. Res.*, **109**, D03206, doi:10.1029/2003JD003914.
- ✓ Yu, H., Y.J. Kaufman, M. Chin, et al. (2006), A review of measurement-based assessments of the aerosol direct radiative effect and forcing, *Atmos. Chem. Phys.*, **6**, 613-666, 2006.
- ✓ Zhang, R., G. Li, J. Fan, D.L. Wu, and M.J. Molina (2007), Intensification of Pacific storm track linked to Asian pollution, *Proc. Natl. Acad. Sci.*, **104**(13), 5295-5299, doi:10.1073/pnas.0700618104.
- ✓ Zhang, X.Y., S.L. Gong, T.L. Zhao, R. Arimoto, Y.Q. Wang, and Z.J. Zhou (2003), Sources of Asian dust and role of climate change versus desertification in Asian dust emission, *Geophys. Res. Lett.*, **30**, 2272, doi:10.1029/2003GL018206

Table 1: Comparisons of MODIS-based estimate and GOCART and GMI simulations of East Asia outflow and North America inflow of pollution aerosol for 2004.

Approach	East Asia Outflow (Tg/year)	North America Inflow (Tg/year)	Inflow/Outflow (%)
MODIS	16.0	4.2	26
GOCART	15.0	4.8	32
GMI	14.4	4.8	34

Table 2: Interannual variations of estimated outflow and inflow pollution fluxes

Year	East Asia Outflow (Tg/yr)	North America Inflow (Tg/yr)	Inflow/Outflow (%)
2002	15.8	3.7	23
2003	20.3	5.7	28
2004	16.0	4.2	26
2005	18.6	4.1	22
2002-2005	17.7	4.4	25

Table 3: Estimated uncertainties in the flux calculation resulting from major assumptions in implementing the algorithm. The overall uncertainty is derived by assuming individual sources of uncertainty are independent.

Major sources of uncertainties	Estimated Uncertainty
Derivation of pollution AOD	50%
Aerosol mass extinction efficiency (dry) MEE	30%
Humidification factor for the extinction $f(RH)$	20%
Using polar-orbiting measurements to represent diurnal average	10%
Transport heights	50%
Overall	80%

Figure Captions:

Figure 1: (a) Illustration of four boxes into which GLAS data are aggregated for deriving average profiles of aerosol backscatter and detection frequency during (b) September-November, 2003, and (c) February-March, 2004.

Figure 2: Schematic of calculating pollution mass fluxes across meridional planes with a width of 10° in longitude in West Pacific (WP) and East Pacific (EP). The latitudinal integration over WP and EP is used to represent East Asia outflow and North America inflow, respectively. The background shows the distribution of pollution aerosol optical depth for MAM 2004.

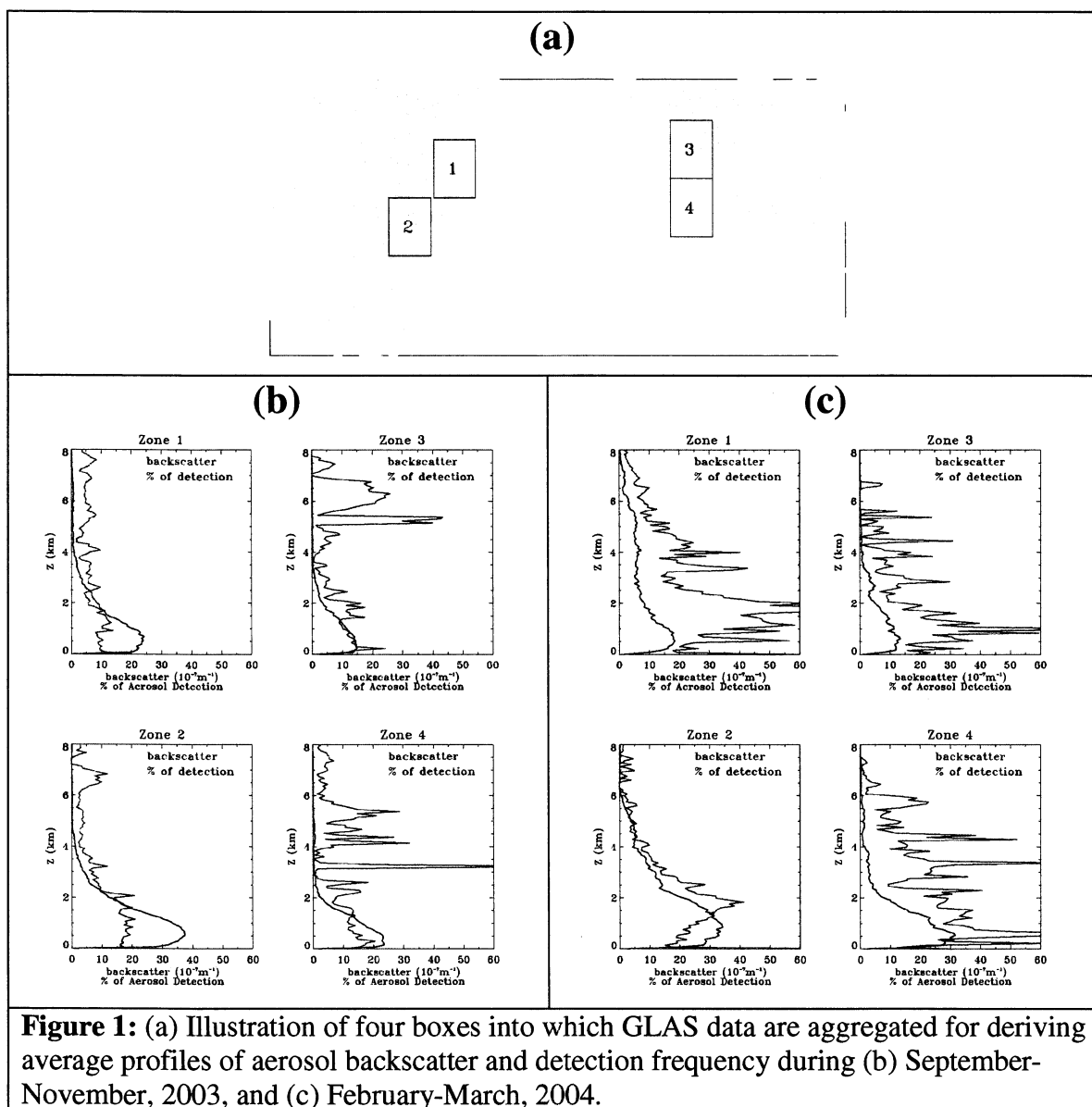
Figure 3: Seasonal variations of MODIS-derived East Asia outflow and North America inflow of pollution aerosol for 2004.

Figure 4: Comparisons of seasonal variations of meridional integrated pollution flux across West Pacific and East Pacific.

Figure 5: Comparisons of meridional variations of annual pollution flux across the West Pacific and East Pacific, respectively.

Figure 6: Interannual variations of pollution aerosol optical depth for (a) MAM, and (b) JJA.

Figure 7: Interannual variations of East Asia outflow pollution aerosol fluxes in different seasons and latitude segments.



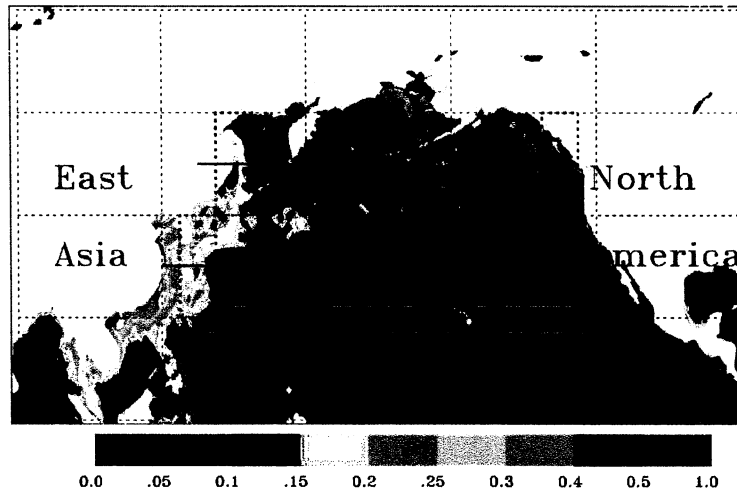


Figure 2: Schematic of calculating pollution mass fluxes across meridional planes with a width of 10° in longitude in West Pacific (WP) and East Pacific (EP). The latitudinal integration over WP and EP is used to represent East Asia outflow and North America inflow, respectively. The background shows the distribution of pollution AOD for MAM 2004.

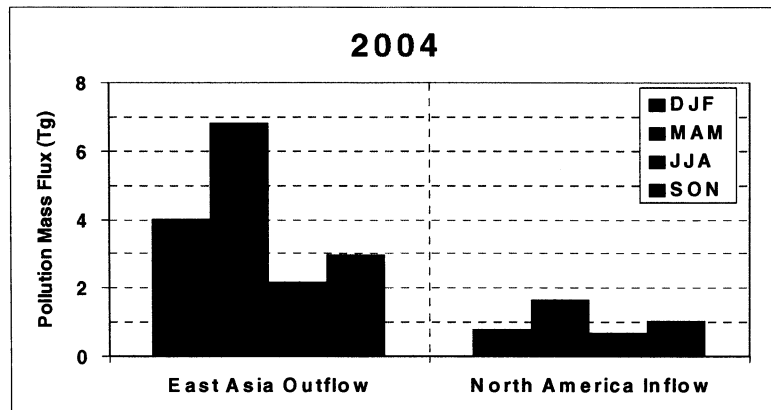
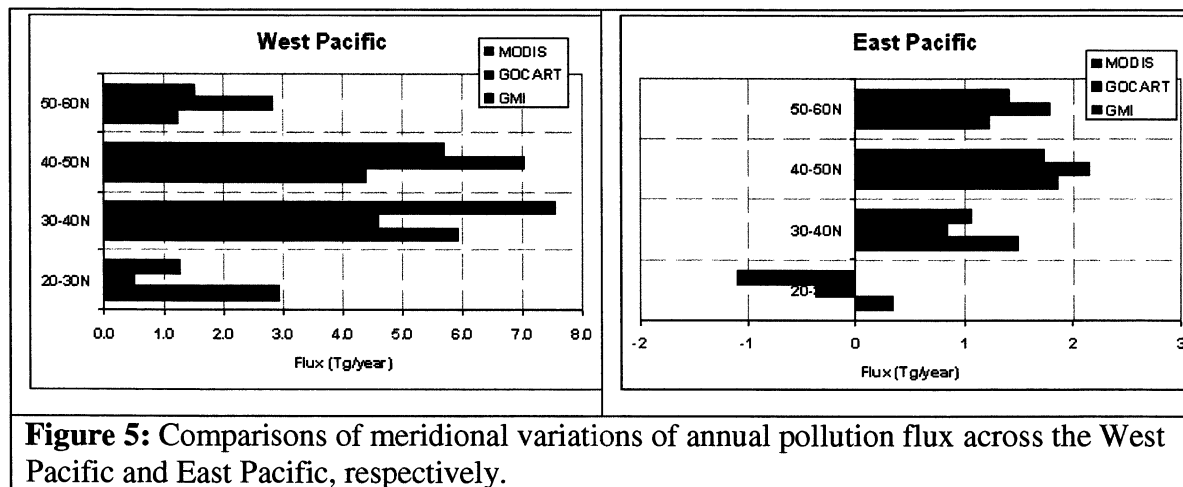
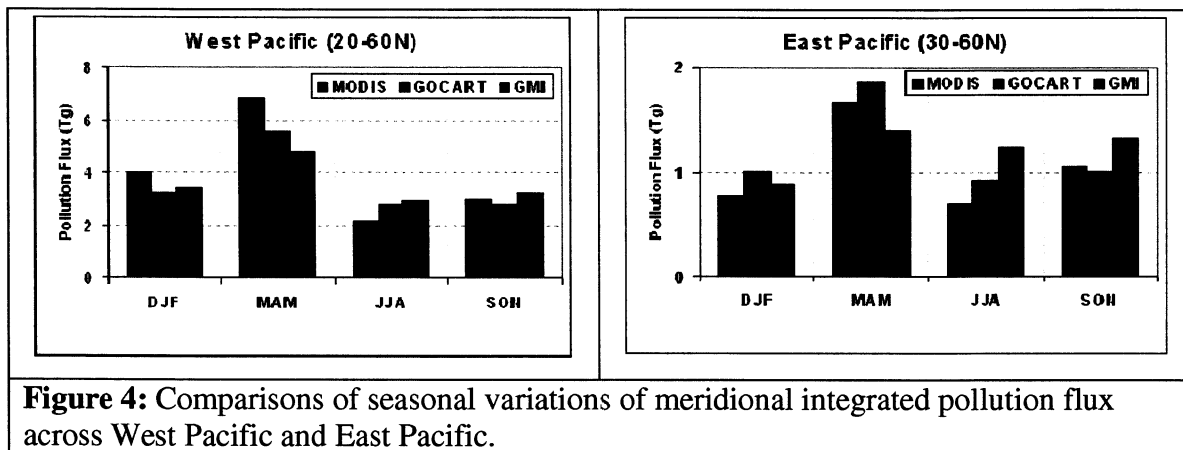
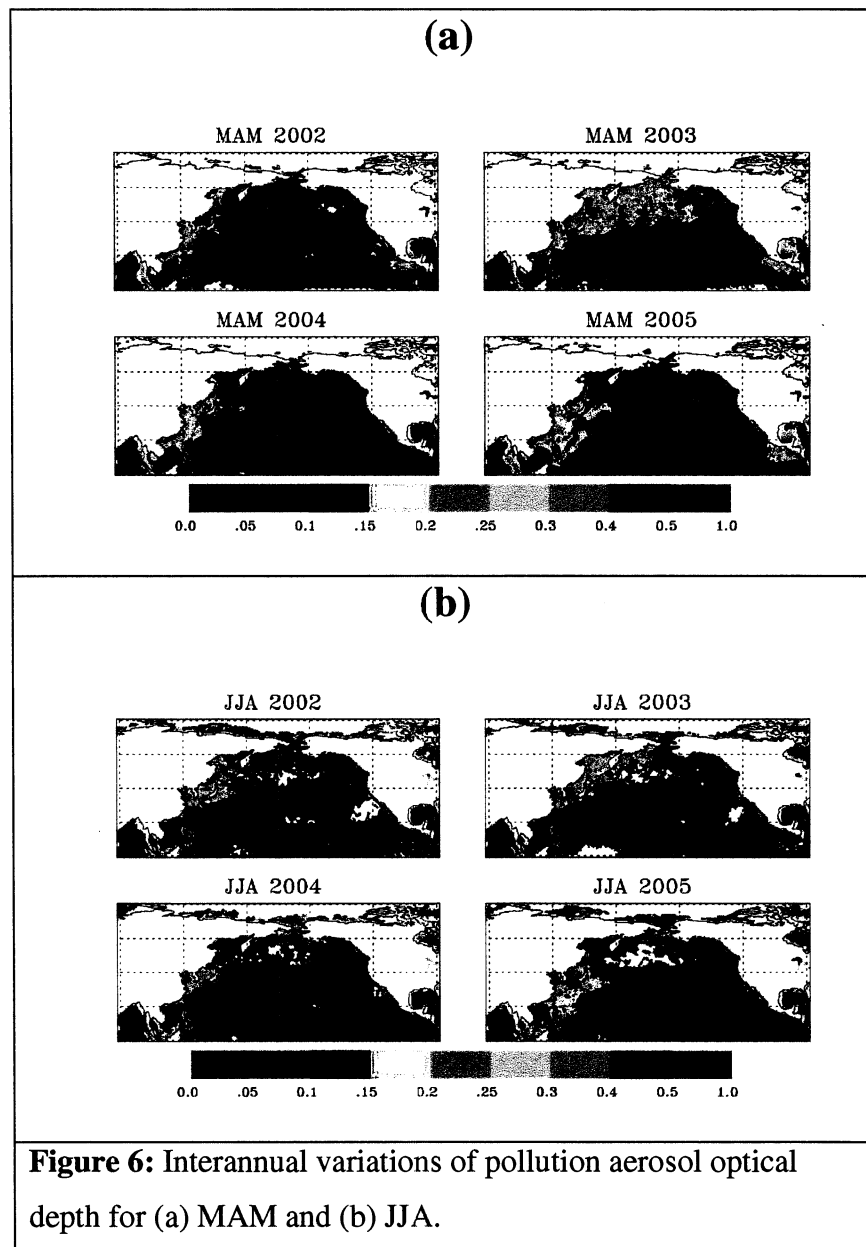


Figure 3: Seasonal variations of MODIS-derived East Asia outflow and North America inflow of pollution aerosol for 2004.





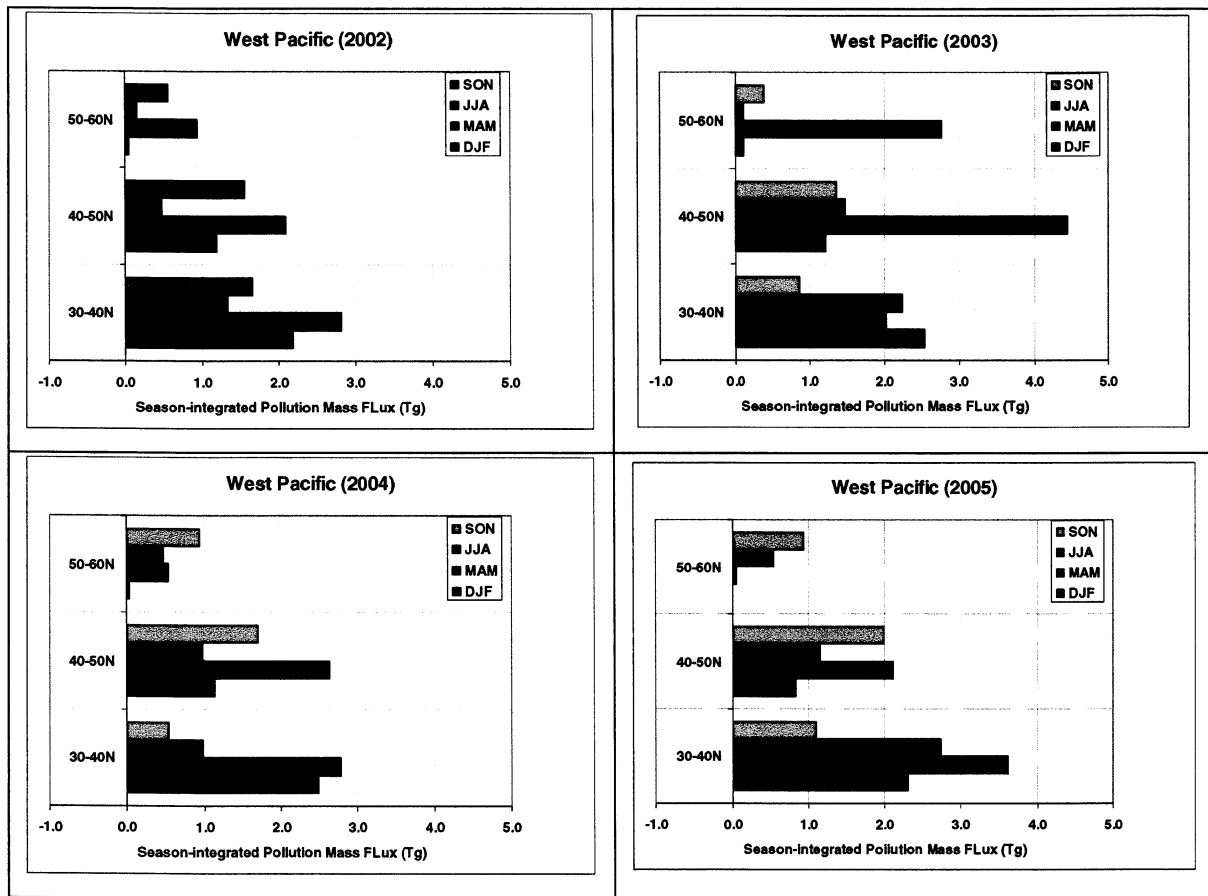


Figure 7: Interannual variations of East Asia outflow pollution aerosol fluxes in different seasons and latitude segments.